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Supplementary Information

Highly uniform monolayer graphene synthesis via facile pretreatment of copper catalyst substrates using an ammonium persulfate solution

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Comparison of other copper surface engineering approaches

Table S1 Comparison of other copper surface engineering approaches

Method	Description	Advantage	Disadvantage	Ref
Surface planarization (Electropolishing)	● Electrochemically etch the copper surface by using it as the anode in the electrolyte in a three-electrode electrochemical cell.	● Produce highly flat copper surface morphology.	● Complicated. ● May be non-uniform producing etch pits.	S1
Surface planarization (Annealing)	● Anneal the Cu samples under sufficient hydrogen flow for several minutes to hours before flowing carbon precursor like methane.	● Produce relatively flat copper surface morphology. ● Reduce native oxide in the copper catalyst which reduces excessive carbon sublimation.	● Usually should be assisted with other surface engineering method to acquire the high quality graphene. ● Could be irregular depending on air flow dynamics in the vacuum chamber. ● Inapplicable in case of low temperature CVD process.	S2, S3
Surface passivation (Melamine)	● Use melamine as passivating agent to suppress the active sites on copper surface.	● Large-sized (~1 cm) single crystal graphene. ● High quality.	● Could be irregular depending on air flow dynamics in the vacuum chamber. ● Because use of the powder form, may be inhaled which is highly toxic. ● Inapplicable in case of low temperature CVD process.	S4
Chemical treatment (Nitric acid)	● Use nitric acid to etch the copper surface for aiding in removing the native oxide on as-received Cu foils in addition to any possible surface contaminants.	● Simple. ● Remove the most of heavy metal particles which induce excessive nucleation.	● Toxic and dangerous etchant. ● Harmful gas byproduct (NO_2) ● May non-uniform depending on the solution-copper contact condition.	S5

Chemical treatment (Acetic acid)	● Use acetic acid to etch the copper surface for aiding in removing the native oxide on as-received Cu foils in addition to any possible surface contaminants.	● Simple.	● Relatively toxic etchant. ● The quality of the grown graphene is usually not good.	S1, S6
<u>Chemical and mechanical treatment (Ammonium persulfate)</u>	● **Use ammonium persulfate solution to etch the copper surface for aiding in removing the native oxide on as-received Cu foils in addition to any possible surface contaminants while applying ultrasonication for uniform etch profile.**	● **Simple.** ● **Uniform surface treatment due to mechanical etching.** ● **No harmful gas byproduct.** ● **Produce good quality of graphene even with rough surface morphology of the copper surface.**	● **Relatively toxic etchant.**	**This work**

Wrinkle density analysis

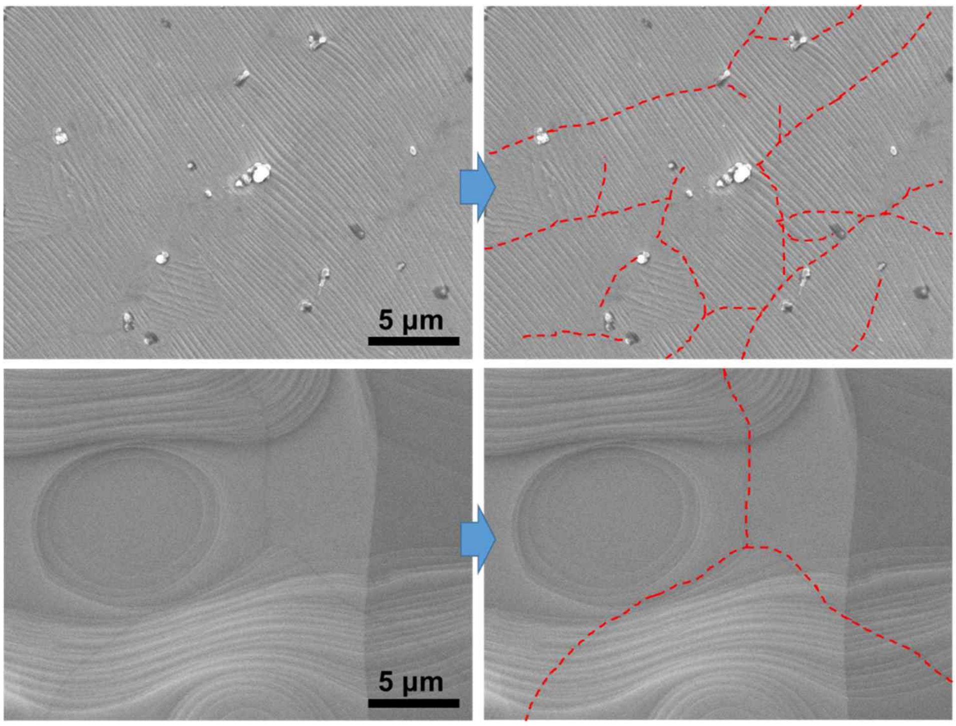

Fig. S1 Upper images: SEM images of the pristine copper surface after graphene growth. Right image shows corresponding wrinkles indicated as red dotted lines. Lower images: SEM images of the cleaned copper surface after graphene growth. Right image shows corresponding wrinkles indicated as red dotted lines. The graphene on the cleaned copper surface exhibits less wrinkle density than that of the pristine one.

High Resolution Transmission Electron Microscopy (HRTEM) characterization of clean graphene

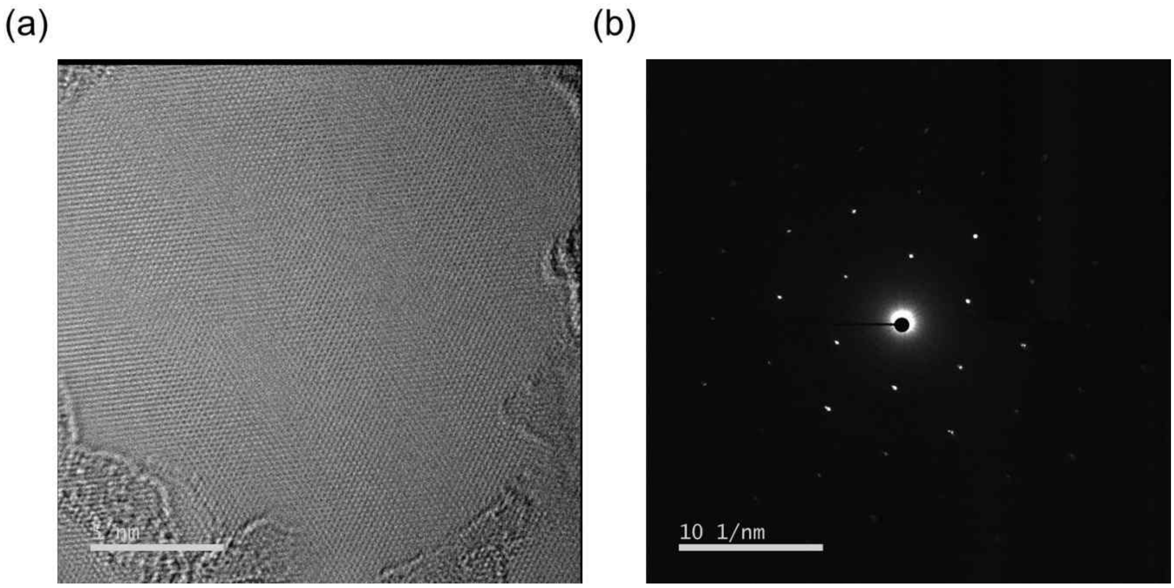

Fig. S2 (a) HRTEM image of clean graphene. (b) Selected Area Electron Diffraction (SAED) image of the clean graphene.

To obtain the HRTEM images, clean graphene were transferred onto Quantifoil TEM grids with 2 μm holes by standard transfer procedures. The accelerating voltage was 60 kV. Fig. S2 (a) shows the bright field HRTEM image of clean graphene. The image supports that the high crystalline structure (i.e. hexagonal honeycomb structure) of the clean graphene. Fig. S2 (b) shows Selected Area Electron Diffraction (SAED) image of the clean graphene, which shows only six-fold nearest reflection spots indicating that the clean graphene is monolayer graphene.

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